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(54) Transparent copolyamides and their use for light guide claddings and moulded bodies

(57) Transparent copolyamides derived from alkyl derivatives of bis(4-aminocyclohexyl)methane and, optionally, other diamines with isophthalic acid and, optionally terphthalic acid or aliphatic dicarboxylic acids and further polyamide forming components having 6 or more carbon atoms, using bis(4-amino-3-methyl-5-ethylcyclohexyl)methane having a selected mixture of isomers which preferably contains trans/trans and/or cis/trans isomers.

The transparent copolyamides are especially suitable for lightwave guide cladding and processing by injection moulding to give moulded bodies.

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SPECIFICATION

Transparent copolyamides and their use for light guide claddings and moulded bodies

The invention relates to new transparent copolyamides based on alkyl derivatives of bis(4-aminocyclohexyl)methane alone or in admixture with other diamines.

Polyamides and copolyamides prepared by the use of 3-aminometyl-3,5,5'-trimethylcyclohexylamine (isophorone diamine, IPD), 4,4-diaminodicylcohexyl or of diamines of the bis(4-aminocyclohexyl)alkane-type, which can be substituted by methyl groups on the cyclohexyl substituents, are known.

The polyamides described in GB-A-0619707 and in US-A-2494563, derived from 4,4'-diaminodicyclohexyl or from diamines of the bis(4-aminocyclohexyl)methane-type and dicarboxylic acids such as adipic acid or sebacic acid are transparent if, in their preparation, the diamines are introduced as liquid isomeric mixtures at 25°C. The workability and certain properties such as the transparence stability and the hydrolysis-resistance with respect to boiling water, the tear resistance in organic solvents of these transparent polyamides, however, leave something to be desired.

The transparent polyamide described in US-A-2696482, derived from isomeric mixtures of 4,4-diaminodicylohexylmethane which are liquid at 25°C and from isophthalic acid has good resistance to hot water. However, in order to conduct the polycondensation satisfactorily, the diphenyl ester of isophthalic acid must be used as a starting material or phenol must be added to the polycondensation mixture as solvent or plasticiser. On account of the high softening temperature and the high melt viscosity of these transparent polyamides, which have a maximum water uptake of 7.75%, working temperatures of about 330°C are necessary.

The transparent polyamide described in US-A-2516585, derived from bis(4-amino-3-methylcyclohexyl)methane and terephthalic acid has similar disadvantages.

The transparent copolyamides described in US-A-3847877, derived from 4,4'-diaminodicylohexylmethane, terephthalic acid and/or isophthalic acid, and caprolactam, have a comparably high capacity for water absorption, and tend to cloudiness after treatment for a few days with boiling water. Further, they contain a proportion of unreacted monomeric caprolactam which reduces or prevents their use in several areas.

This last feature applies also to the copolyamides described in DE-A-1595354, derived from 2,2-bis(4-aminocyclohexyl)propane, dicarboxylic acids with more than 20% w/w caprolactam and/or a further conventional polyamide-forming components, e.g. hexamethylenediammonium adipate, which are soluble in methanol.

In so far as the transparent polyamides described in DE-A-1595354 are prepared from 2,2-bis(4-aminocyclohexyl)propane and a dicarboxylic acid such as adipic acid alone, they have better solvent resistance, but have poor workability on account of their high softening point. It is not possible to prepare from them injection mouldings free from strain.

The transparent copolyamides described in US-A-3597400, derived from 4,4'-diaminodicylohexylmethane, hexamethylenediamine, terephthalic acid and isophthalic acid, have a high water absorption capacity. As a result, on storing these transparent copolyamides in water, their softening points are reduced to 50–60°C.

The transparent copolyamides described in US-A-3842045 which are polycondensation products of 4,4'-diaminodicyclohexylmethane which is only 40–54% in the trans-trans configuration, and a mixture of 50–70 mol% 1,10-decanedicarboxylic acid and 30–50 mol % suberic acid or azelaic acid, contain neither aromatic dicarboxylic acid nor an additional polyamide-forming component. This is also the case for the transparent copolyamides derived from 2-2-bis(4-aminocyclohexyl)propane and/or its methyl derivatives and from dicarboxylic acid mixtures which comprise 20–65 mol % adipic acid and 35–80 mol % suberic acid, azelaic acid, sebacic acid and/or 1,10-decanedicarboxylic acid, according to the teaching of US-A-3840501.

CH-A-0449257 also relates to transparent polyamides derived from 1,10-decanedicarboxylic acid and diamines of the dicycane type, including bis(4-amino-3-methylcyclohexyl)methane or 2,2-bis(4-aminocyclohexyl)propane with similar structure.

DE-A-2405985 discloses flame-repellent, thermoplastic moulding materials containing red phosphorus and also a transparent polyamide or mixtures of two or more transparent polyamides. Among the transparent polyamides, those which are named include those derived from 35 mol % 4,4'-diaminodicyclohexylmethane or 2,2-bis(4-aminocyclohexyl)propane, 35 mol % isophthalic acid and 30 mol % ω-aminolauric acid (or its corresponding lactam) or a stoichiometric mixture of dodecamethylenediamine and 1,10-decanedicarboxylic acid. The monomer mixtures used for the preparation of these polyamides comprise 33 or 31.5% w/w ω-aminolauric acid or the stoichiometric mixture of dodecamethylenediamine and 1,10-decane-dicarboxylic acid, respectively.

DE-A-2936759 describes transparent copolyamides having a high glass transition point, into which at least 30% w/w of a ω-aminocarboxylic acid having at least 11 C atoms are introduced

in order to reduce the high processing viscosity, and which contains a considerable amount of isophoronediamine in addition to a diamine of the dicycane type. However, this leads to brittleness and discolouration of the copolyamides.

EP-A-0012931 describes transparent copolyamides which are derived from, inter alia, adipic acid, hexamethylenediamine and, as further diamine component, a mixture of bis(4-aminocyclohexyl)methane. In consequence, the hot water resistance is insufficient.

DE-A-2642244 describes transparent copolyamides derived from ω-aminocarboxylic acid, isophthalic acid and a methyl derivative of bis(4-aminocyclohexyl)methane, which exhibit unsatisfactory heat moulding resistance, strain crack corrosion, toughness, transparence stability and hydrolysis-resistance in boiling water, and have the disadvantage of a relatively high working viscosity.

US-A-4293687 describes the general class of bis(4-aminocyclohexyl)methane-type diamines, and their preparation. The disclosure of this patent specification is directed towards certain crystalline linear polyamides and not to amorphous copolyamides.

The preparation of the 3,5-diethyl homologue of bis(4-aminocyclohexyl)methane, described in GB-A-2170209, is relatively expensive. Owing to the low reactivity of these diamine components, high temperatures and long reaction times are necessary for the preparation of the copolyamides described therein, in order to obtain satisfactory viscosity.

The invention has the object of producing easily processable copolyamides having high transparency and very good properties in use, in particular for use as a lightwave conductor cladding and for injection moulding to moulded bodies, which no longer have the disadvantages, described above, of the known copolyamides. Surprisingly, it has been found that an asymmetrically-substituted bis(4-aminocyclohexyl)methane is of especial value, and which has reactivity dependent on its isomeric form.

According to the invention, new transparent copolyamides are obtainable by polycondensation of alkyl derivatives of bis(4-aminocyclohexyl)methane and, optionally, further diamines, with isophthalic acid and, optionally, terephthalic acid or aliphatic dicarboxylic acids and further polyamide-forming components having 6 or more carbon atoms, characterised in that a selected mixture of isomers of bis(4-amino-3-methyl-5-ethylcyclohexyl)methane, which preferably contains trans/trans and/or cis/trans isomers, is used as the alkyl derivative of bis(4-aminocyclohexyl)methane.

According to a specific embodiment, the transparent copolyamides according to the invention are prepared by polycondensation of

a) bis(4-amino-3-methyl-5-ethylcyclohexyl)methane as a selected mixture of isomers, which preferably contains trans/trans and/or cis/trans isomers, alone or in admixture with other diamines, with

b) the approximately stoichiometric amount, with respect to components a), of isophthalic acid, of which 0–50% (mol or by weight) can be replaced by terephthalic acid or its homologues or 5 to 98% w/w by further aliphatic dicarboxylic acids, and

c) 20 to 60% by weight of the sum of a), b) and c), of one or more polyamide-forming components such as C1) a ω-aminocarboxylic acid or its lactam having more than 8 C atoms and/or C2, a salt or 1 : 1 stoichiometric mixture of a dicarboxylic acid, in particular a α,ω-polymethylenedicarboxylic acid and an aliphatic diamine, in particular a α,ω-polymethylenediamine, wherein the condition applies that there is an average number of at least 7 methylene groups in c) per amide group or per pair of amide-forming groups, and the mininum number of methylene groups between the amide-forming groups is 6, and wherein, further, when part of the isophthalic acid is replaced by an aliphatic dicarboxylic acid, the sum of the weights of the aliphatic dicarboxylic acids, and the additive c) must lie in the range of 20–60% by weight of the sum of a), b) and c).

Highly transparent copolyamides are thus obtained on the addition of the polyamide-forming components according to c), and which have good processability, very good mechanical properties and excellent transparence stability and hydrolysis-resistance in boiling water, as well as a good tensile strength with respect to organic solvents, and remain transparent even in blends with other polyamides, especially with nylon 11 and 12.

Nowhere in the available state of the art is there any indication of the use of the steric influences on the reactivity of the amino groups of the bis(4-aminocyclohexyl)methane derivative which is introduced, not only in terms of the nature and position of the alkyl substituents, but also in connection with the isometry of the cyclohexyls. The selected isomeric mixture of the bis(4-amino-3-methyl-5-ethylcyclohexyl)methane according to a) therefore means a selected reactive mixture which contains only low amounts, preferably below 20%, and most especially less than 10% w/w, of cis/cis isomers. The proportion of the trans/trans and cis/trans isomers is then at least 80%, but preferably more than 90% w/w. It is apparently important that the 3-methyl and 5-ethyl groups are each in the immediate vicinity of the amino group.

The further diamine components which, if desired, are used in addition to the alkyl derivative of bis(4-aminocyclohexyl)methane according to the invention, preferably comprise bis(4-amino-3-

methylcyclohexyl)methane, bis(4-amino-3,5-diethylcylohexyl)methane, bis(4-amino-3-methyl-5-isopropylcyclohexyl)methane, bis(4-aminocyclohexyl)methane, 2,2-bis(4-aminocyclohexyl)propane or further substituted diamines of the bicyclohexylalkane-type, 1,3-bis(aminomethyl)cyclohexane, 3-aminomethyl-3,5,5-trimethylcyclohexylamine, 2,4,4-(2,2,4)-trimethylhexamethylenediamine, hexamethylenediamine, methylpentamethylenediamine, 3(4),8(9)-bisaminomethyltricylo[5,2,1,$0^{8,6}$]decane or 2,6-bis(aminometyl)norbornane-m-xylylenediamine.

The bis(4-amino-3-methyl-5-ethylcyclohexyl)methane can be mixed with such diamines in the mol ratio 0.9:0.1 to 0.1:0.9. However, for such additional diamines, it is not necessary to make any restrictions with respect to the existing isomers.

For use as the acid components according to b), isophthalic acid alone or mixtures of isophthalic acid and terephthalic acid which contain up to 50% (mol or weight) terephthalicacid and/ substituted isophthalic acids are suitable. 5 to 98% of the isophthalic acid can be replaced by aliphatic dicarboxylic acids. Such dicarboxylic acids are advantageously those having more than 6 C atoms, especially suberic acid, azelaic acid, sebacic acid, decanedicarboxylic acid, undecanedicarboxylic, dodecanecarboxylic acid, tridecanedicarboxylic acid, and their side chain-substituted homologues.

Polyamide-forming components according to C1 include laurolactam or ω-aminolauric acid and ω-aminoundecanoic acid or a mixture thereof. Those according to C2 include salts of α,ω-diaminoalkanes such as 1,6-diaminohexane, 1,8-diamnooctane, 1,9-diaminononane, 1,10-diaminodecane, 1,12-diaminododecane, 1,13-diaminotridecane or 2,2-4- or 2,4,4-trimethylhexamethylenediamine, and α,ω-dicarboxylic acids such as azelaic acid, sebacic acid, decanedicarboxylic acid or dodecanedicarboxylic acid, and substituted homologues thereof.

If several compounds or salt pairs are used according to C1 and C2, the condition applies that the average number of the methylene groups in c) per amide group is at least 7. $-NH_2$ and $-COOH$ are to be understood as amide-forming groups.

The equivalent weight is the same as the molecular weight for compounds of the type C1. For salts or stoichiometric mixtures of diamine and dicarboxylic acids of the type C2, it is half the sum of the weight of the dicarboxylic acid and the diamine.

The starting materials for the copolyamides according to the invention are especially suitable for polycondensation in the melt. They are temperature-stable and are not very likely to dicolour during polycondensation, even if temperatures up to 330°C are used and longer reaction times than usual are appropriate.

The copolyamides prepared according to the invention have glass transition temperatures of about 110 to about 190°C and high heat moulding resistance, and exhibit outstanding toughness, transparence and resistance in boiling water against chemicals and very favourable processing viscosities.

It is particularly advantageous to use an amount of c) so that the glass transition temperature is in the range of 130–180°C. By reduction of the added amount the glass transition temperature rises, and vice versa.

By comparison with the transparent copolyamides disclosed in US-A-3842045, US-A-3840501 and DE-A-2405985, the copolyamides prepared according to the invention have a higher transparence stability in boiling water.

By comparison with the copolyamides described in DE-A-2642244, the polyamides of the invention have higher heat moulding stability, lower strain tear corrosion in alcoholic solvents, greater troughness, lower working viscosity and, further, better transparence and resistance in boiling water or, as the case may be, organic solvents. By comparison with the polyamides described in GB-A-2170209, the products according to the invention can be prepared in shorter reaction times at lower temperatures, and exhibit higher heat moulding stability, higher impact resistance and transparency and improved flow properties, e.g. in the form of a tool.

By the addition of the starting materials according to the invention, it is possible to reach a melt viscosity of less than 2000 Pa.s at 270°C and a load of 122.6 N, which allows problem-free processability in the preparation of moulded bodies and on the extrusion of very small dimensional parts.

In general, known methods of polycondensation are used in the preparation of copolyamides according to the invention. The diamines and the dicarboxylic acids must be used in equivalent amounts, in order that copolyamides with the desired molecular weights are obtained.

Using controlled addition of excesses, usually of diamine but also of dicarboxylic acid, the chain length of the copolyamides can be regulated without the introduction of monoamines or monocarboxylic acids. The components according to a) and b) can be added as such or also as salts.

ω-aminoundecanoic acids can be added directly to the reaction mixture as component C1). In a preferred embodiment, however, instead of ω-aminolauric acid, its lactam, laurolactam, is used. This requires that a pressure reaction phase in the presence of water is conducted into order to split the lactam ring before the desired polycondensation.

If, according to C2), a dicarboxylic acid and a diamine are used, they can be added alone or in

the form of their salts. Salts of straight-chain, α,ω-dicarboxylic acids and α,ω-diamines can be produced relatively easily. Their use involves no problems of stoichiometry.

In the condensation of the diamines according to a) with isophthalic acid, with a mixture of isophthalic acid and terephthalic acid or a substituted isophthalic acid or an aliphatic dicarboxylic acid and with laurolactam, the mixture of the starting materials (which still contains water) is first subjected to pressure at elevated temperature. Following pressure release, the water is removed during polycondensation under an inert gas (usually nitrogen) or vacuum.

If diamines and dicarboxylic acids alone are used, the neutralisation reaction proceeds, with the addition of some water, at temperatures at which a stirrable mixture or a melt exists, and the temperature is raised gradually. In order that there is no loss of amine, pre-condensation can be conducted in closed systems under pressure; after release of pressure, further polycondensation can be conducted without pressure or under vacuum.

Conventional additives in the preparation of polyamides can be added to the polycondensation mixture before, during or towards the end of the polycondensation; these additives should advantageously be soluble in the copolyamide, for the purposes of transparency. These additives are, for example, antioxidants, flame-retardants, light stabilisers, platicisers, mould separation agents, optical brighteners and pigments. Such additives can be mixed with the copolyamide or worked in by re-melting in suitable apparatus, e.g. in an extruder.

The copolyamides according to the invention are very suitable for processing in so-called injection moulding or extrusion processes, e.g. for the preparation or cladding of optical guides and also for the most various moulded bodies.

A further advantage of the copolyamides according to the invention is that, depending on the melt viscosity of the relevant granulate, they can be used without danger of discoloration at injection temperatures up to 310°C and higher. The material exhibits good flow and deformation properties. In order to improve the mould filling, the tools are tempered, so that the deformability and also the transparency can be influenced positively. A further advantage is that conventional pulverisation of the granulate with certain slip agents, before further processing, is very economical.

The copolyamides according to the invention can also be alloyed or mixed with other homo-or copolyamides or mixtures thereof or with other plastics materials. For example, this can be achieved by mixing and extruding the granulate or plastics components.

Examples of additional homopolyamides are nylon 12, nylon 11, nylon 6.6, nylon 6.9, nylon 6.10; examples of copolyamides are those which contain the monomers which give the specified homopolyamides, or other copolyamides; examples of other polymers are those which are at least partially compatible with the copolyamides according to the invention.

These co-components are advantageously added in an amount of 0–50% with respect to the resulting alloy. The alloying of a further component with copolyamides prepared according to the invention can change the mechanical properties; for example, the impact strength and notch toughness are generally improved, or the stiffness reduced, thereby.

A further advantage of the copolyamides of the invention is that, if further additives according to c), for example monomers of nylon 12, are used, the transparence stability in boilding water is substantially unaffected.

The following Examples 1 to 11 and 14 to 17 illustrate the invention. Examples 12 and 13 are comparative. In all the Examples (except 5), the diamine bis(4-amino-3-methyl-5-ethylcyclohexyl)methane was used in the form of a selected liquid isomeric mixture (85% trans/trans and cis/trans proportion) as component a).

Example 1 to 11 and Comparative Example 12 and 13

The following Table I sets out the conditions used in the various polycondensation tests, in which only one diamine component was used as component a). * indicates that a change from the general procedure was made (see below).

Column 2 gives the nature of component c), and column 3 the proportion (% w/w) of component c) based on the total weight of all components a), b) and c). Column 4 gives the equivalent (mol) ratio of the components a), b) and c). Column 5 gives the period and column 6 the maximum reaction temperature.

Results are given in Table III: The viscosity ηrel was measured as a 0.5% w/w solution in m-cresol at 20°C. A DSC 990 apparatus from DuPont (R=5/E, S=20°C/min) was used for the T_G measurement. The melt viscosity (η melt) was measured (in Pa.s) with a melt index testing apparatus Goettfert M/21.6 (nozzle length 8 mm, diameter 2.1 mm) at 270°C and a load of 122.6 N. For the measurement of the transparence stability in boiling water, the copolyamide was prepared as platelents and tested in boiling water; Very good=transparence stability of several weeks/good=transparence stability of about 3 days/average=transparence stability of about 1 day/bad=transparence stability of only a few hours.

The bending E modulus was measured for small bodies according to DIN 53,452 which have been prepared on a laboratory injection moulding machine. Stress crack stability was measured

for test bodies (127 × 12.7 × 3.2 mm) in 100% ethanol: the value gives the peripheral fibre stress in N/mm² after 90 seconds working-in of the ethanol for a test body maintained under a mechanical load.

In Example 5, a bis(4-amino-3-methyl-5-ethylcyclohexyl)methane having a high content (40%) of cis/cis isomers, is used.

In Example 9, component b), otherwise isophthalic acid, was replaced by tert-butylisophthalic acid. In Example 10, 25% of the isophthalic acid were replaced by terephthalic acid. In Example 11, 1,10-decanedicarboxylic acid was introduced instead of isophathalic acid.

The components were weighed into a 4 litre condensation apparatus made of steel, which was carefully flushed with nitrogen before and after the introduction. The apparatus was heated carefully to 200°C under nitrogen, with good stirring of the mixture of starting materials. Pre-condensation is thus induced, and the predominant proportion of the water of reaction distilled into a receiver.

The melt thus became increasingly viscous. The temperature was then gradually increased, and reached 280–300°C after a further hour. After a total condensation period of 4–8 hours, pressure was released and the melt was finely passed into a cold water bath through a bottom valve: the solidified strands were pulverised, using a comminutor, to a granulate which was then dried under vacuum.

Examples 12 and 13 were conducted in accordance with the disclosure of DE-A-2642444. Both copolyamides thus prepared have higher melt visosities, lower bending E moduli and lower stress crack stability than tests of Examples with copolyamides according to the invention.

Examples 14 to 16

Table II sets out tests in which a second diamine is used in addition to bis(4-amino-3-methyl-5-ethylcyclohexyl)methane as component a). Isophthalic acid was always used as component b) and ω-aminolauric acid as component c). The results are given in Table III.

TABLE I

Examples	c)	c) (% w/w)	Equivalence ratio a) : b) : c) (equiv.)	Time (h)	max. react. temperature (°C)
1	ω-Amino-lauric acid	32.6	1.06 : 1 : 1.03	4.0	280
2	Aminolauric acid	32.6	1.06 : 1 : 1.03	3.0	280
3	ω-Aminoundec-anoic acid	29.3	1 : 0.9 : 1.0	4.5	280
4	9,12-Salt	47	1 : 0.95 : 1.0	6	280
5	ω-Amino-lauric acid	30	1* : 0.92 : 1.0	5.5	280
6	ω-Amino-lauric acid	32	1 : 1 : 1	6	280
7	ω-Amino-lauric acid	33	1 : 0.98 : 1.1	6.5	280
8	ω-Amino-lauric acid	30.5	1 : 0.95 : 1.0	6	285
9	ω-Amino-lauric acid	30.7	1.06 : 1* : 1.04	7	280
10	ω-Amino-lauric acid	30.7	1 : 0.9 : 1	7	275
11	ω-Amino-lauric acid	27	1 : 1.14* : 1.2	5.5	280
12	ω-Amino-lauric acid	36.5	1 : 1 : 1	5.5	285
13	ω-Amino-lauric acid	33.8	1 : 0.99 : 1.05	4.5	285

TABLE II

Example	Second diamine	Mol ratio of the two diamines	a) : b) : c) (equiv.)
14	Bis(4-amino-cyclohexyl)-methane	50 : 50	1.16 : 1.0 : 1.06
15	Hexamethylene-diamine	86 : 24	1 : 0.97 : 0
16	Bis(4-amino-3-methyl-cyclo-hexyl)methane	50 : 50	1 : 0.98 : 0.90

TABLE III

Example	ηrel	T_G (°C)	η melt (Pa.s)	Transparency in boiling water	Bending E Modulus	Crack formation (N/mm²)
1	1.56	162	2024	very good	-	-
2	1.586	163	2084	very good	-	-
3	1.40	150	880	good	2100	> 12
4	1.56	122	1080	very good	2320	> 12
5	1.31	130	497	bad	1800	≧ 9
6	1.515	146	2089	very good	2129	> 15
7	1.524	162	1941	good	2060	> 13
8	1.39	167	1640	very good	2100	> 12
9	1.377	172	476	good	2520	> 11
10	1.46	150	960	good	2380	> 12
11	1.51	146.5	1220	good	1944	-
12	1.68	156	2146	good	1632	> 2.5
13	1.52	154	1900	good	1816	> 4
14	1.56	148	1260	very good	-	-
15	1.47	138	392	good	-	-
16	1.541	156	1982	very good	-	-

Example 17

This Example is intended to demonstrate the ability to prepare copolyamides according to the invention in a semi-technical procedure.

16.7 kg of the liquid isomeric mixture of bis(4-amino-3-methyl-5-ethylcyclohexyl)methane containing 85% w/w trans/trans and cis/trans isomers, 11.5 kg laurolactam, 1.0 g silicone-based anti-foaming agent, 10 kg water and finally 9.4 kg isophthalic acid were introduced into a 150 l V45A polycondensation autoclave. The autoclave was flushed several times with nitrogen and, after closing, gradually heated to 180°C until a clear melt was obtained. Stirring ensued, at about 100 rpm, and the temperature was raised to 290°C. There was thus a pressure in the autoclave of about 18 bar, which was maintained for 2 hours. Condensation under N_2 for 2.5 hours at 280°C followed. Finally, the melt was removed from the autoclave as a cord and comminuted to granulate. After drying, 42 μ equivalent/g carboxyl groups and 18 μ equivalent/g amino groups, a ηrel of 1.52, η melt of 1260 Pa.s (270°C/122.6 N) and a T_G of 163°C were measured.

Small DIN beams and DIN tensile rods were obtained from an injection moulding machine at a bulk temperature of 290°C, and on which various mechanical properties were measured.

In an impact resistance test according to DIN 53453, the test bodies did not break, the limiting bending tension according to DIN 53452 was 125 N/mm², the bending E modulus 2322 N/m².

The rods exhibited very good hydrolysis and transparence properties in boilding water, and a tension-corrosion-tear resistance of more than 15 N/mm² after immersion in 100% ethanol for 90 seconds.

The equivalent weight water uptake at 23°C (50% atmospheric pressure was only 1.2%. The dimensional stability of sprayed moulded bodies of the copolyamide according to the invention was excellent.

Sample granulate of this preparation was co-extruded with 25 and 30% w/w polyamide 12 chips. The extrudate thus-obtained was fully transparent and had T_G values of 102°C to 90°C.

CLAIMS

1. A transparent copolyamide obtainable by polycondensation of a diamine component comprising bis(4-amino-3-ethyl-5-methlcyclohexyl)methane with isophthalic acid and, optionally, terephthalic acid or aliphatic dicarboxylic acids, and further polyamide-forming components having 6 or more C atoms.

2. A copolyamide according to claim 1, wherein the bis(4-amino-3-methyl-5-ethylcyclohexyl)-methane is predominantly in trans/trans and/or cis/trans form.

3. A transparent copolyamide according to claim 1 or claim 2, wherein the diamine component comprises a further diamine, and the molar ratio of the two dimaines is 0.9:0.1 to 0.1:0.9.

4. A copolyamide according to claim 3, wherein the further diamine is selected from bis(4-amino-3-methylcyclohexyl)methane, bis(4-amino-3,5-diethylcyclohexyl)methane, bis(4-amino-3-methyl-5-isopropylcyclohexyl)methane, bis(4-aminocyclohexyl)methane, 2,2-bis(4-aminocyclohexyl)-propane, 1,3-bis(aminomethyl)cyclohexane, 3-aminomethyl-3,5,5-trimethylcylohexylamine, 2,4,4-(2,2,4)-trimethylhexamethylenediamine, 3(4),8(9)-bis(aminomethyl)tricyclo[5.2.1.$0^{2,6}$]decane, 2,6-bis(aminomethyl)norbornane and m-xylenediamine.

5. A copolyamide according to any preceding claim, wherein the acid component comprises isophthalic acid alone, a mixture of isophthalic and terephthalic acids containing up to 50% (mol or by weight) terephthalic acid, or a mixture of 2–95% w/w isophthalic acid and 98–2% w/w aliphatic dicarboxylic acid.

6. A copolyamide according to any preceding claim, wherein the further polyamide-forming components having 6 or more C atoms form 20 to 60% wt. of the total amount of the transparent copolyamide and comprise ω-aminocarboxylic acids or their lactams having more than 8 C atoms and/or a salt or a stoichiometric 1:1 mixture of an aliphatic dicarboxylic acid and an aliphatic diamine, wherein the condition applies that there is an average number of at least 7 methylene groups per amide group or per pair of amide-forming groups, and the minimum number of methylene groups between the amide-forming groups is 6, and wherein, further, when part of the isophthalic acid is replaced by an aliphatic dicarboxylic acid, the sum of the weights of these aliphatic dicarboxylic acids and the said polamide-forming components must lie in the range of 20–60% by weight, based on the total amount of the transparent copolyamide.

7. A polyamide according to claim 6, wherein the aliphatic dicarboxylic acid is an α,ω-polymethylene-dicarboxylic acid and the aliphatic diamine is an α,ω-polymethylenediamine.

8. A copolyamide according to any preceding claim, wherein the bis(4-amino-3-methyl-5-ehtylcyclohexyl)methane consists of at least 80% trans/trans and/or cis/trans isomers.

9. A copolyamide substantially as described in any of Examples 1 to 11 and 14 to 17.

10. A mixture of a copolyamide according to any preceding claim with another polyamide.

11. A mixture according to claim 10, wherein the other polyamide is polyamide 11 or 12 or a copolyamide thereof.

12. A cladding material, for opto-electronic conducting systems, which comprises a product according to any preceding claim.

13. A moulded body prepared from a product according to any of claims 1 to 11.

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